

This Page Is Inserted by IFW Operations
and is not a part of the Official Record

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images may include (but are not limited to):

- BLACK BORDERS
- TEXT CUT OFF AT TOP, BOTTOM OR SIDES
- FADED TEXT
- ILLEGIBLE TEXT
- SKEWED/SLANTED IMAGES
- COLORED PHOTOS
- BLACK OR VERY BLACK AND WHITE DARK PHOTOS
- GRAY SCALE DOCUMENTS

IMAGES ARE BEST AVAILABLE COPY.

**As rescanning documents *will not* correct images,
please do not report the images to the
Image Problem Mailbox.**

431/4

PCT

WORLD INTELLECTUAL PROPERTY ORGANIZATION
International Bureau



INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

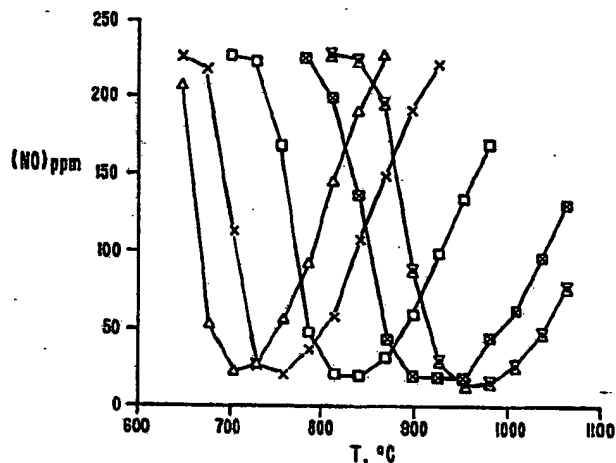
(51) International Patent Classification 5 : C01B 21/00		A1	(11) International Publication Number: WO 93/19006
			(43) International Publication Date: 30 September 1993 (30.09.93)
(21) International Application Number: PCT/US93/02391 (22) International Filing Date: 17 March 1993 (17.03.93) (30) Priority data: 07/858,316 25 March 1992 (25.03.92) US (71) Applicant: ENERGY & ENVIRONMENTAL RESEARCH CORP. [US/US]; 18 Mason, Irvine, CA 92718 (US). (72) Inventors: HO, Loc ; 1014 N. Mayflower Street, Anaheim, CA 92801 (US). CHEN, Shih, L. ; 14 Alameda, Irvine, CA 92720 (US). SEEKER, William, Randall ; 711 Calle Brisa, San Clemente, CA 92672 (US). MALY, Peter, M. ; 25261 Tanoak Lane, El Toro, CA 92630 (US).		(74) Agents: SEELEY, David, O. et al.; Workman, Nydegger & Jensen, 1000 Eagle Gate Tower, 60 East South Temple, Salt Lake City, UT 84111 (US). (81) Designated States: CA, PL, European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE). Published <i>With international search report.</i>	

(54) Title: **METHOD FOR CONTROLLING N₂O EMISSIONS AND FOR THE REDUCTION OF NO_x AND SO_x EMISSIONS IN COMBUSTION SYSTEMS**

(57) Abstract

Methods for controlling N₂O emissions from stationary combustion systems having variable flue gas temperatures are disclosed. The N₂O emissions are controlled by the introduction of an N₂O control agent, such as an alkaline compound, into the effluent stream. In addition, the present invention discloses methods for controlling N₂O emissions from stationary combustion systems having variable flue gas temperatures while reducing NO_x emissions. Use of an NO_x reducing agent and an N₂O control agent, such as urea and monosodium glutamate, enlarges the temperature window for effective selective noncatalytic NO_x reduction while significantly eliminating N₂O emissions commonly experienced with urea injection. Further, the present invention discloses methods for controlling N₂O emissions from stationary combustion systems having variable flue gas temperatures while reducing SO_x emissions. Use of an NO_x reducing agent, an SO_x reducing agent, and an N₂O control agent, such as urea, an alkaline-earth metal compound, and an alkali-metal compound, respectively, enlarges the temperature window for effective NO_x and SO_x reductions while significantly eliminating N₂O emissions. The figure is a graph illustrating how hydrogen mixed with ammonia shifts the temperature window of the selective non catalytic NO_x reduction process to lower temperatures.

Reduction of NO by H₂/NH₃ mixtures



(NO)₀ = 225 ppm, mass flow through reactor constant and corresponding to 0.1s at 1038 °C

x for 0-H₂/NH₃

m for 0.125

□ for 0.50

x for 1.3

△ for 2.4

(NH₃)₀ / (NO)₀ = 2

FOR THE PURPOSES OF INFORMATION ONLY

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AT	Austria	FR	France	MR	Mauritania
AU	Australia	GA	Gabon	MW	Malawi
BB	Barbados	GB	United Kingdom	NL	Netherlands
BE	Belgium	GN	Guinea	NO	Norway
BF	Burkina Faso	GR	Greece	NZ	New Zealand
BG	Bulgaria	HU	Hungary	PL	Poland
BJ	Benin	IE	Ireland	PT	Portugal
BR	Brazil	IT	Italy	RO	Romania
CA	Canada	JP	Japan	RU	Russian Federation
CF	Central African Republic	KP	Democratic People's Republic of Korea	SD	Sudan
CG	Congo	KR	Republic of Korea	SE	Sweden
CH	Switzerland	KZ	Kazakhstan	SK	Slovak Republic
CI	Côte d'Ivoire	LJ	Liechtenstein	SN	Senegal
CM	Cameroon	LK	Sri Lanka	SU	Soviet Union
CS	Czechoslovakia	LU	Luxembourg	TD	Chad
CZ	Czech Republic	MC	Monaco	TG	Togo
DE	Germany	MG	Madagascar	UA	Ukraine
DK	Denmark	ML	Mali	US	United States of America
ES	Spain	MN	Mongolia	VN	Viet Nam
FI	Finland				

METHOD FOR CONTROLLING N_2O EMISSIONS AND FOR THE REDUCTION OF NO_x
AND SO_x EMISSIONS IN COMBUSTION SYSTEMS

5

BACKGROUND

1. The Field of the Invention

10 The present invention is directed to an improved process for controlling N_2O emissions in combustion systems. More particularly, the present invention is directed to methods for controlling N_2O emissions by introducing a N_2O control agent into the effluent stream. In addition, the processes of the present invention significantly eliminate
15 N_2O emissions while providing a wide temperature window for effective NO_x reduction by the use of an NO_x reducing agent. Further, the processes of the present invention significantly eliminate N_2O emissions while enhancing SO_x reduction by the use of an SO_x reducing agent.

20

2. Technology Overview

The combustion of fossil fuels, industrial wastes, and municipal wastes produces significant amounts of NO_x and SO_x which ultimately participate in the formation of photochemical smog and acid rain. In addition to NO_x and
25 SO_x , the combustion and incineration processes often produce another class of pollutants, namely, nitrous oxides (commonly referred to as " N_2O "). N_2O is a potential "greenhouse" gas and can also lead to degradation of the stratospheric ozone layer.

30

There exist many processes for the control of NO_x and SO_x emissions from power plants and incinerators. However, control of N_2O emissions from incinerators is a problem to which no presently available technology provides a fully
35 satisfactory answer. While the amount of N_2O produced by burning fuel or waste can be minimized by managing the combustion process, waste or fuel typically contains substantial amounts of chemically bound nitrogen such that N_2O levels are usually unacceptably high, even with careful control of the combustion process. As a result, some form
40 of post-combustion N_2O control technology must be used.

Whereas the control of N_2O emissions is critical, the control of another class of pollutants - nitrogen oxides

(commonly referred to as "NO_x") -is also important, and current methods of reducing NO_x emissions result in the formation of high N₂O emissions. While the amount of NO_x produced by burning fuel or waste can be minimized by
5 managing the combustion process, as with N₂O emissions, waste typically contains substantial amounts of chemically bound nitrogen such that NO_x levels are usually unacceptably high, even with careful control of the combustion process. As a result, some form of post-combustion technology must
10 be used which not only controls N₂O emissions, but NO_x emissions as well.

Two types of post-combustion NO_x control technologies are presently available, selective catalytic reduction ("SCR") and selective noncatalytic reduction ("SNCR").
15 Applications of SCR to incinerators are generally regarded as nonfeasible because waste contains virtually all possible trace impurities and these impurities can act as catalyst poisons.

As a result, SNCR processes have been adopted as the
20 best available NO_x control technology for incinerators. In the usual SNCR process, a nitrogen-containing reducing agent, normally either ammonia (NH₃) or urea (H₂NCONH₂), is contacted with flue gas within a relatively narrow temperature range. The optimum contacting temperature is
25 typically a factor of the reducing agent. A homogeneous gas phase reaction occurs which reduces the NO_x in the flue gas to molecular nitrogen (N₂) and water (H₂O).

In most applications, the performance of the NO_x reduction processes depends primarily on the available
30 reaction time, i.e., the length of time the flue gas spends in the temperature range suitable for NO_x reduction by the chosen reducing agent. For applications in which the available reaction time is less than 0.2 seconds, NO_x reductions in the 60% to 80% range are typically achieved.
35 For applications in which the available reaction time is greater than 0.2 seconds NO_x reductions in the 80% to 90% range have commonly been achieved.

The design of a modern incinerator provides the post-flame gases with a residence time generally greater than

1.0 seconds in the temperature range appropriate to NO_x reduction processes. Hence, one might expect incinerators to be a very favorable application for selective noncatalytic NO_x reduction. Instead, however, NO_x reduction in incinerators is typically only 40% or even less.

The poor performance of NO_x reduction processes on incinerators is, in part, a result of the fact that the temperature of the flue gas in incinerators is more highly variable than it is in other combustion systems. Waste is inherently a fuel with a highly variable BTU content. This variability causes the temperature of the flue gases downstream of the combustion zone to be nonhomogeneous in space and to fluctuate in time.

If the temperature of the flue gas is a little too low at the point where the reducing agent is injected, slight or no NO_x reduction occurs. If the temperature is too high, the nitrogen-containing reducing agent (NH₃ or H₂NCONH₂) has some tendency to oxidize to produce NO_x, and the net reduction of NO_x is poor or more NO_x may even be produced. Because this range of temperatures ("temperature window") for the NO_x reduction process is narrow, successful application of the process is always critically dependent on locating the reducing agent injection system at the location at which the average temperature is optimum for the process.

In any application, however, the temperature will be nonhomogeneous, and process performance will be determined by an average over a temperature range. Since this always includes some temperatures which are too high and some which are too low for good NO_x reduction, the practical extent of NO_x control which the process can provide is always significantly less than is achieved in laboratory experiments.

Since the range of the NO_x reduction temperature window increases with increasing reaction time, the longer reaction time available in incinerators compensates, in part, for this difficulty. However, there is an additional problem: the optimum temperature for NO_x reduction may be shifted. For example, as shown in Figure 1, (quoted from

R.K. Lyon and J.E. Hardy, "Discovery and Development of the Thermal DeNO_x Process," 25 Ind. Eng. Chem. Fundam. 19 (1986); see also 21 Environmental Science and Technology, 232, (1987)) hydrogen (H₂) mixed with the ammonia shifts the NO_x reduction temperature window to lower temperatures. The magnitude of the temperature shift increases as the amount of H₂ is increased.

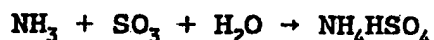
This shifting of the temperature window is a general effect which occurs with other combustible materials, including CO, natural gas, etc. Even though the temperature window may be shifted, the size of the temperature window is not enlarged to a significant degree by the presence of other reducing agents in the combustion effluent stream.

Whereas some reducing agents of NO_x emissions, which enlarge the useful temperature window for NO_x reduction, are known in the art, certain problems may result in their use. While certain of the agents disclosed in this art reduce NO_x emissions, the formation of N₂O emissions is also experienced as discussed previously. Thus, benefits incurred by some agents which can reduce NO_x emissions is negatively countered if those agents act to replace the NO_x emissions with an alternate pollutant such as N₂O emissions.

The poor performance of N₂O and NO_x reduction processes also encounters a severe disadvantage due to the amount of sulfur contained in the fuel to be incinerated. In most instances, it is quite expensive and technically difficult to remove the sulfur before burning the fuel. It has been a common practice, therefore, to burn fuel which still contains a portion of the native sulfur. Sulfur compounds, including SO₂ and SO₃, are then produced during combustion and must be removed from the effluent gas stream by separate expensive technologies.

Combustion of a sulfur-containing fuel in a boiler, furnace, or other burner typically produces combustion effluents in which 98% to 99% of the sulfur exist in the form of SO₂, and only 1% to 2% of the sulfur exists in the form of SO₃. In processes where NO is reduced by injecting ammonia into the gas stream, some of the ammonia or ammonia

precursors injected into the process will be left unreacted. Under certain conditions, NH_3 will react with the sulfur gases, i.e., as the combustion effluents cool, the remaining NH_3 will react with SO_3 and water vapor present in the effluent stream to form NH_4HSO_4 according to reaction equation (1);



Unfortunately, NH_4HSO_4 is an extremely sticky and corrosive liquid and is known to damage the equipment used in combustion processes.

The temperature at which the formation of NH_4HSO_4 occurs is such that, in a typical boiler or furnace of the type generally encountered in combustion processes, formation of NH_4HSO_4 occurs within the air heater. Thus, fouling, corrosion, and plugging of the air heater has been commonly encountered. As a result, the nitric oxide reduction technologies which use ammonia and ammonia precursors have had severe limitations and their commercial acceptance has been correspondingly limited. The operators of boilers and furnaces are often extremely reluctant to accept any technology which can cause fouling, plugging, and corrosion such as that expected from NH_4HSO_4 .

It will be noted that in the event that there are no sulfur oxides in the effluent stream, the formation of NH_4HSO_4 is not a problem. In many combustion applications, however, there will be at least a small quantity of sulfur oxides in the effluent stream and, thus, some SO_3 . The ammonia or ammonia precursors which are used in the nitric oxide reduction reactions can then react with the SO_3 present to form NH_4HSO_4 , which even in small quantities can cause severe problems over time. Moreover, any SO_x emissions which do not form NH_4HSO_4 , may react with water to form a mist of sulfuric acid. This can also cause severe problems over time.

From the foregoing, it is apparent that what is currently needed in the art are methods for controlling N_2O

emissions from stationary combustion systems having variable flue gas temperatures.

It would also be an improvement in the art if the present invention could provide methods for controlling N_2O emissions from stationary combustion systems while enlarging the useful temperature window and enhancing NO_x reduction.

It would be a further advancement in the art to provide methods for controlling N_2O emissions from stationary combustion systems which allows one to treat flue gas with ammonia or ammonia precursors to reduce NO_x emissions without producing an alternate class of pollutants such as N_2O .

It would be a still further advancement in the art to provide methods for controlling N_2O emissions from stationary combustion systems while enhancing SO_x reduction.

It would be a still further advancement in the art to provide methods for controlling N_2O emissions from stationary combustion systems while enlarging the useful temperature window for NO_x and SO_x reduction.

It would be an additional advancement in the art to provide methods for controlling N_2O emissions from stationary combustion systems which are economically feasible.

It would also be an additional advancement in the art to provide methods for controlling N_2O emissions from stationary combustion systems which perform effectively.

Such methods for controlling NO_x emissions from stationary combustion systems are disclosed and claimed herein.

BRIEF SUMMARY OF THE INVENTION

The present invention relates to methods for controlling N_2O emissions from stationary combustion systems. The present invention provides a process for controlling N_2O in emissions in effluent streams by introducing an N_2O controlling agent into the effluent stream within a certain temperature window. Preferably, the N_2O control agent is an alkaline compound. This process

significantly eliminates N_2O emissions from the combustion of the fuel.

Moreover, the present invention relates to methods for controlling N_2O emissions while reducing NO_x emissions from stationary combustion systems having variable flue gas temperatures. The present invention provides a process for controlling N_2O emissions while reducing NO_x emissions in effluent streams by introducing an NO_x reducing agent and an N_2O control agent into the effluent stream within a certain temperature window. Preferably, the NO_x reducing agent is ammonia or an ammonia precursor. The N_2O control agent is an alkaline compound, as defined at a later point. This process substantially reduces or eliminates the adverse effects of variable flue gas temperatures so as to reduce NO_x emissions while significantly eliminating N_2O emissions. The elimination of N_2O emissions is necessary because the injection of urea or ammonium/urea to effluents in stationary combustion systems to control NO_x emissions results in the production of higher N_2O emissions.

The present invention also provides a process for reducing SO_x emissions (which may or may not be present in effluent streams containing NO_x emissions) by introducing an SO_x reducing agent, and an N_2O control agent into the effluent stream within a certain temperature window. Preferably, the SO_x reducing agent is an alkaline earth-metal compound. The N_2O control agent is an alkaline compound. This process substantially reduces or eliminates the adverse effects of variable flue gas temperatures so as to reduce SO_x emissions while significantly eliminating N_2O emissions.

The present invention is further applicable to controlling N_2O emissions while reducing NO_x and SO_x emissions in effluent streams by introducing an NO_x and SO_x reducing agent, and an N_2O control agent into the effluent stream within a certain temperature window. Preferably, the NO_x reducing agent is ammonia or an ammonia precursor. The SO_x reducing agent is an alkaline-earth metal compound. The N_2O control agent is an alkaline compound, as described below. This process substantially reduces or eliminates

the adverse effects of variable flue gas temperatures so as to reduce NO_x and SO_x emissions while significantly eliminating N_2O emissions.

5

BRIEF DESCRIPTION OF THE DRAWINGS

In order to better understand the manner in which the above-recited and other advantages and objects of the invention are obtained, a more particular description of the invention briefly described above will be rendered by reference to a specific embodiment thereof which is illustrated in the appended drawings. Understanding that these drawings depict only a typical embodiment of the invention and are not therefore to be considered limiting of its scope, the invention will be described and explained with additional specificity and detail through the use of the accompanying drawings in which:

Figure 1 is a graph illustrating how hydrogen mixed with ammonia shifts the temperature window of the selective noncatalytic NO_x reduction process to lower temperatures.

Figure 2 is a graph comparing NO_x reduction efficiency of urea and urea plus an amount of monosodium glutamate.

Figure 3 is a graph comparing N_2O emissions of urea and urea plus an amount of monosodium glutamate.

Figure 4 is a graph illustrating N_2O reduction efficiency of aqueous Na_2CO_3 .

Figure 5 is a graph illustrating SO_x reduction efficiency by the addition of alkali metals to limestone.

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENT

The present invention relates to methods for controlling N_2O emissions from stationary combustion systems such as power plant boilers, process furnaces, and incinerators. Additionally, the present invention relates to methods for controlling N_2O emissions while reducing NO_x emissions from stationary combustion systems which have variable flue gas temperatures, the N_2O emissions being produced by the treatment of NO_x with ammonia or ammonia precursors. Moreover, the present invention relates to methods for controlling N_2O emissions while reducing SO_x

emissions from stationary combustion systems. The present invention also relates to methods for controlling N_2O emissions while reducing NO_x and SO_x emissions from stationary combustion systems.

5 Specifically, the present invention is directed to a process for controlling N_2O in effluent streams having temperature variations and containing N_2O and excess oxygen. The process comprises the step of introducing an N_2O control agent into the effluent stream at a point where the
10 temperature of the effluent stream is in the range from about 1300°F to about 3000°F. Preferably, the process comprises the step of introducing an N_2O control agent into the effluent stream at a point where the temperature of the effluent stream is in the range from about 1800°F to about
15 2800°F.

The N_2O control agent of the process is an alkaline compound. The term "alkaline compounds" as used in this specification is meant to refer to compounds which comprise any of the univalent, mostly basic metals of Groups I and
20 II of the periodic table. These basic metals comprise lithium, sodium, potassium, rubidium, cesium, francium, magnesium and calcium. The alkaline compounds generally comprise alkaline-metal compounds of an organic acid, alkali-metal hydroxides, alkali-metal carbonates, alkaline-earth metal compounds, and the like.
25

Some specific examples of alkaline compounds which fall within the scope of the present invention include sodium-based compounds ($NaOH$, Na_2SO_4 , $NaHSO_4$, and Na_2CO_3), $Mg(CH_3COO)_2$ or $Ca(CH_3COO)_2$. Another example is monosodium
30 glutamate which is the monosodium salt of the naturally occurring L-form of glutamic acid. The compound is manufactured by the fermentation of carbohydrate sources. Monosodium glutamate is very soluble in water, and sparingly soluble in alcohol. It must be remembered, however, that
35 the present invention is not limited to these specific examples, and that any compound fitting within and/or exhibiting the properties of alkaline compounds as broadly defined may also be included.

In one embodiment of the present invention, the N_2O control agent is an alkali-metal compound of an organic acid. Preferably, the N_2O control agent is an alkali-metal compound of an organic acid such as sodium glutamate, potassium glutamate, sodium formate, potassium formate, sodium acetate, and potassium acetate. Nevertheless, it should be understood that, while the foregoing may be preferred under certain conditions, the present invention is not limited to the foregoing group of compounds.

In another embodiment of the present invention, the N_2O control agent is an alkali-metal hydroxide. Preferably, the N_2O control agent is an alkali-metal hydroxide selected from the group consisting essentially of LiOH, NaOH, and KOH. Nevertheless, it should be understood that the present invention is not limited to the foregoing group of compounds.

In still another embodiment of the present invention, the N_2O control agent is an alkali-metal carbonate. Preferably, the N_2O control agent is an alkali-metal carbonate selected from the group consisting essentially of lithium carbonate, lithium bicarbonate, sodium carbonate, sodium bicarbonate, potassium carbonate, and potassium bicarbonate. Nevertheless, it should be understood that the present invention is not limited to the foregoing group of compounds.

In yet another embodiment of the present invention, the N_2O control agent is an alkaline-earth metal compound. Preferably, the N_2O control agent is an alkaline-earth metal compound selected from the group consisting essentially of CaO , $CaCO_3$, $Ca(OH)_2$, $Ca(COOH)_2$, $Ca(CH_3COO)_2$, MgO , $MgCO_3$, $Mg(OH)_2$, $Mg(COOH)_2$, and $Mg(CH_3COO)_2$. Nevertheless, it should be understood that the present invention is not limited to the foregoing group of compounds.

An examination of the equivalent m le fraction of the alkali and/or alkaline-earth metals in the N_2O control agent can also be determined. Preferably, the equivalent m le fraction of the alkali and/or alkaline-earth metals in the N_2O control agent is in the range from about 5 to about 2000 ppm by volume, in the effluent stream. Most preferably,

the N_2O control agent is in the range from about 50 ppm to about 200 ppm by volume, in the effluent stream.

5 The amount of N_2O emissions that can be expected following the use of the N_2O control agent are in the range from about 0 ppm to about 50 ppm. Preferably, the limited N_2O emissions which do occur in the range from about 0 ppm to about 10 ppm.

10 As previously indicated, the present invention can be successfully used to significantly suppress the formation of N_2O in flue gases having a temperature in the range from about 1300°F to about 2800°F at the point of contacting. With regard to the N_2O control agent, the reaction time is preferably greater than 0.1 seconds and more preferably greater than 0.2 seconds. The temperature decay versus
15 time in the reaction zone of the present invention is preferably less than about 600°F/second, and most preferably less than about 200°F/second.

It should also be mentioned that the oxygen concentration in the N_2O containing effluent streams shall
20 be in the range from about 0.1% to about 15% by volume of oxygen to total flue gas. Preferably, the oxygen concentration in the effluent streams is in the range from about 2% to about 9%. It should be noted that the term "controlling agent," as used throughout this application,
25 is meant to illustrate that there are limited amounts, if any, of N_2O emissions. Controlling can mean that the N_2O control agent suppresses the formation of N_2O emissions. Controlling can also mean that the N_2O control agent significantly eliminates N_2O emissions.

30 The present invention is also directed to a process for controlling N_2O emissions while reducing NO_x in effluent streams having temperature variations and containing NO_x and excess oxygen. The process comprising the step of
35 introducing an NO_x reducing agent into an effluent stream at a point where the temperature of the effluent stream is in the range from about 1300°F to about 2000°F, preferably in the range from about 1600°F to about 1900°F.

The process also comprises the step of introducing an N_2O control agent into the effluent stream at a point where

the temperature of the effluent stream is in the range from about 1300°F to about 3000°F, preferably in the range from about 1800°F to about 2800°F. The N₂O control agent of this process is similar to the N₂O control agent employed in the process for controlling N₂O emissions.

The NO_x reducing agent of the process is ammonia and/or an ammonia precursor, such as urea, ammonium sulfate, ammonium carbonate, ammonium bicarbonate, ammonium formate, ammonium acetate, and ammonium oxalate. Nevertheless, it should be understood, however, that the NO_x reducing agent is not limited to the foregoing group of compounds, but would include other compounds capable of reducing NO_x emissions.

Preferably, it has been found that by using a combination of urea and alkaline compounds, over a range of ratios, effective NO_x reduction can be obtained over a significantly wider temperature range than by using conventional noncatalytic NO_x reduction processes with ammonia, urea, or a combination of ammonia and urea. In this way, the adverse effects of variable flue gas temperatures can be substantially reduced or eliminated and the increased reduction of NO_x emissions can occur while the control over the N₂O emissions is maintained.

Within the instant invention, the NO_x reducing agent and the N₂O control agent may be injected into the flue gas as a mist in a single step. Preferably, urea and alkaline compound are injected into the flue gas in a single step. The introduction of the compounds in a single step can be differentiated from a step-wise or multi-stage process wherein a plurality of treatment fluids are injected into a combustion effluent stream at a plurality of temperature zones. It can be understood that, not only does the single stage use of urea and alkaline compound provide for effective NO_x and SO_x reductions over a significantly wider temperature range, but also that a single stage step is less complicated and less expensive.

Preparation of the solution containing urea and an alkaline compound is accomplished in many different ways. In one embodiment of the process for reducing NO_x, the N₂O

control agent and the NO_x reducing agent are mixed together and injected at the same location. Preferably, the urea and an alkaline compound are premixed in a predetermined ratio as a solution and injected in a single stage into the flue gas. Alternatively, solutions containing the urea and an alkaline compound, are impingently mixed in a single stage into the flue gas.

Preferably, urea and alkaline compounds are typically purchased in a solid form. Their use as a solution above, therefore, requires that the solids be dissolved in some type of a solvent. Examples of solvents include water and alcohol; however, it should be understood that other solvents, known to those skilled in the art, exist which will dissolve urea and/or an alkaline compound.

In the preferred embodiment of the present invention, an aqueous solution of urea is prepared, the concentration of urea being in the range from about 2% to about 30%. Thereafter, the aqueous urea and alkaline compound solution is injected as a mist into the combustion effluent stream.

Alternatively, in another embodiment, the N_2O control agent and the NO_x reducing agent are injected at separate locations. The concentration of the urea solution is in the range from about 2% to about 30%. The concentration of the alkaline compound is in the range from about 0.25% to about 7%.

The presence of an NO_x reducing agent in the flue gas is in a predetermined stoichiometric ratio. The amount of NO_x reducing agent, or specifically the urea, in the effluent stream can also be determined by examining the molar ratio of nitrogen in the NO_x reducing agent to the NO_x in the effluent stream. Preferably, the molar ratio of nitrogen in the NO_x reducing agent to the NO_x in the effluent stream is in the range from about 0.5:1 to about 5:1. Most preferably, the molar ratio of nitrogen in the NO_x reducing agent to the NO_x in the effluent stream is in the range from about 1:1 to about 3:1.

An examination of the equivalent mole fraction of the alkali and/or alkaline-earth metals in the N_2O control agent can also be determined. Preferably, the equivalent mole

fraction of the alkali and/or alkaline-earth metals in the N_2O control agent is in the range from about 5 to about 2000 ppm by volume, in the effluent stream. Most preferably, the N_2O control agent is in the range from about 50 to about 200 ppm by volume, in the effluent stream.

It has also been found that the injection of an NO_x reducing agent and an N_2O control agent into a flue gas provides for the significant elimination of N_2O emissions in the flue gas. This is contrary to the injection of pure urea or other urea combinations into flue gas which actually produces high N_2O emissions.

The amount of N_2O emissions that can be expected following the use of urea and an alkaline compound when compared to N_2O emission formed in the presence of urea alone are in the range from about 0 ppm to about 20 ppm. Preferably, the limited N_2O emissions which do occur in the range from about 0 ppm to about 5 ppm.

As discussed above, it can be appreciated that the term "controlling agent," as used throughout this application, is meant to illustrate that there are limited amounts, if any, of N_2O emissions. Controlling can mean that the N_2O control agent suppresses the formation of N_2O emissions. Controlling can also mean that the N_2O control agent significantly eliminates N_2O emissions.

As previously indicated, the present invention can be successfully used to reduce NO_x in flue gases having a temperature in the range from about 1300°F to about 2000°F at the point of contacting. With regard to the introduction of the N_2O control agent and the NO_x reducing agent, the reaction time is preferably greater than 0.1 seconds and more preferably greater than 0.2 seconds. The temperature decay versus time in the reaction zone of the present invention is preferably less than about 600°F/second, and most preferably less than about 200°F/second.

The present invention can also be successfully used to significantly suppress the formation of N_2O in flue gases having a temperature in the range from about 1300°F to about 2800°F at the point of contacting. With regard to

the N_2O control agent and the NO_x reducing agent, the reaction time is preferably greater than 0.1 seconds and more preferably greater than 0.2 seconds. The temperature decay versus time in the reaction zone of the present invention is preferably less than about 600°F/second, and most preferably less than about 200°F/second.

It should also be mentioned that the oxygen concentration in the N_2O and NO_x containing effluent streams shall be in the range from about 0.1% to about 15% by volume of oxygen to total flue gas. Preferably, the oxygen concentration in the effluent streams is in the range from about 2% to about 9%. The present invention is further applicable to controlling N_2O emissions while reducing SO_x emissions in effluent streams having temperature variations and containing SO_x and excess oxygen. It should be understood that in some instances, N_2O emissions occur in conjunction with SO_x emissions and not in the presence of NO_x emissions.

The process comprises the step of introducing an SO_x reducing agent into an effluent stream at a point where the temperature of the effluent stream is in the range from about 800°F to about 3000°F, preferably, in the range from about 1800°F to about 2800°F. The process further comprises the step of introducing an N_2O control agent into the effluent stream at a point where the temperature of the effluent stream is in the range from about 1300°F to about 3000°F, preferably, in the range from about 1800°F to about 2800°F.

The SO_x reducing agent of the process is an alkaline-earth metal compound. Preferably, the SO_x reducing agent is an alkaline-earth metal compound selected from the group consisting essentially of CaO , $CaCO_3$, $Ca(OH)_2$, $Ca(COOH)_2$, $Ca(CH_3COO)_2$, MgO , $MgCO_3$, $Mg(OH)_2$, $Mg(COOH)_2$, and $Mg(CH_3COO)_2$. Nevertheless, it should be understood that the present invention is not limited to this specific group of compounds.

Generally, the steps taken with regard to the SO_x reducing process are similar to the NO_x reducing process. Specifically, the N_2O control agent and the SO_x reducing

agent are mixed together and injected at the same location or the N_2O control agent and the SO_x reducing agent are injected at separate locations. Also, the molar ratio of the alkaline-earth metal in the SO_x reducing agent to the SO_x in the effluent stream is in the range from about 0.5:1 to about 5:1. Additionally, the equivalent mole fraction of the alkali metal in N_2O control agent is in the range from about 10 to about 2000 ppm, by volume, in the effluent stream. Further, the oxygen concentration in said effluent streams is in the range from about 0.1% to about 15%, preferably, in the range from about 2% to about 9%.

The present invention is also directed to a process for controlling N_2O emissions while reducing NO_x and SO_x in effluent streams having temperature variations and containing NO_x , SO_x , and excess oxygen. The control of pollutants in effluent streams naturally requires that in some instances one would want to control the presence of each of these three types of pollutants.

As discussed above, existing selective noncatalytic NO_x reduction processes control NO_x emissions from stationary combustion systems by injecting nitrogen-containing reducing agents such as ammonia or ammonia precursors (e.g., urea) either alone or in combination with a second reducing agent, into the flue gases. The nitrogen-containing reducing agent causes a homogeneous gas phase reaction to occur which reduces NO_x to molecular nitrogen and water. Unfortunately, selective noncatalytic NO_x reduction processes that employ ammonia or urea are ineffective since they lead to the unfavorable production of N_2O emissions. Moreover, as discussed above, the use of reducing agents such as ammonia or urea to treat flue gases can lead to the undesirable formation of NH_4HSO_4 production due to the presence of SO_x emissions. Thus, there may be instances where one must control the presence of N_2O , NO_x , and SO_x emissions.

The process comprises the step of introducing an NO_x reducing agent into an effluent stream at a point where the temperature of the effluent stream is in the range from about 1300°F to about 2000°F, preferably, in the range from

about 1600°F to about 1900°F. The process also comprises the step of introducing an SO_x reducing agent into an effluent stream at a point where the temperature of the effluent stream is in the range from about 800°F to about 3000°F, preferably, in the range from about 1800°F to about 2800°F. The process further comprises the step of introducing an N₂O control agent into the effluent stream at a point where the temperature of the effluent stream is in the range from about 1300°F to about 3000°F, preferably, in the range from about 1800°F to about 2800°F.

Generally, the steps taken with regard to the NO_x and SO_x reducing process is similar to the NO_x and the SO_x reducing processes discussed above. Specifically, the NO_x reducing agent, SO_x reducing agent, and N₂O control agent of this process is similar to the N₂O control agent employed in the processes for reducing NO_x and SO_x emissions. In addition, the molar ratio of nitrogen in the NO_x reducing agent to the NO_x in the effluent stream is in the range from about 0.5:1 to about 5:1; the molar ratio of the alkaline-earth metal in the SO_x reducing agent to the SO_x in the effluent stream is in the range from about 0.5:1 to about 5:1; the equivalent mole fraction of the alkali metal in N₂O control agent is in the range from about 5 to about 2000 ppm, by volume, in the effluent stream; and the oxygen concentration in said effluent streams is in the range from about 0.1% to about 15%.

One distinction which must be made is in regard to the introduction of the different agents in this process. In one embodiment, the NO_x reducing agent, the N₂O control agent, and the SO_x reducing agent are mixed together and injected at the same location. In another embodiment, the NO_x reducing agent, the N₂O control agent, and the SO_x reducing agent are injected at different locations. In still another embodiment, the NO_x reducing agent and the N₂O control agent are injected at the same location but separate from the injection location of the SO_x reducing agent. In a further embodiment, the SO_x reducing agent and the N₂O control agent are injected at the same location but separate from the injection location of the NO_x reducing

agent. In a still further embodiment, the SO_x reducing agent and the NO_x reducing agent are injected at the same location but separate from the injection location of the N_2O control agent.

5 Experiments were conducted to determine the performance of the N_2O control agent in eliminating N_2O emissions. Experiments were also conducted to determine the performance of the N_2O control agent in eliminating N_2O emissions while reducing NO_x emissions. Experiments were
10 also conducted to determine the performance of the N_2O control agent in eliminating N_2O emissions while reducing SO_x emissions.

 The following examples illustrate the suppression of the formation of N_2O emissions achieved by the present
15 invention. The following examples further illustrate the improved NO_x reduction achieved by the present invention over a wide temperature range in addition to eliminating N_2O emissions. The following examples further illustrate the improved SO_x reduction achieved by the present invention
20 over a wide temperature range in addition to eliminating N_2O emissions. These examples are intended to be, purely exemplary of the use of the invention and should not be viewed as limiting the scope of the present invention.

25 Example 1

 In this example, the NO_x reduction efficiency (NO_i/NO_f) of urea/monosodium glutamate and urea were compared over a given temperature range. A 1×10^6 BTU/hour furnace fired with natural gas at 3.5% excess oxygen was used to conduct
30 the experiment. The initial NO_x concentration (" NO_i ") in the combustion effluents ranged from about 297 to about 303 ppm. The molar ratio of nitrogen in reducing agent to NO_i was 1.5. The temperature decay versus time in the reaction zone was approximately 400°F/second. The experimental
35 results are reproduced in Table 1 and shown graphically in Figure 2.

Table 1

	<u>Chemical</u>	<u>NO_i (ppm)</u>	<u>N/NO_i</u>	<u>Peak Temp. (°F)</u>	<u>NO_i/NO_i</u>
5	Urea	298	1.50	1454	1.0
	Urea	299	1.50	1614	0.86
	Urea	303	1.50	1720	0.36
	Urea	297	1.50	1847	0.32
10	Urea	298	1.50	2040	0.83
<hr/>					
	Urea + MSG	298	1.50	1454	0.5
	Urea + MSG	299	1.50	1614	0.13
15	Urea + MSG	303	1.50	1720	0.12
	Urea + MSG	297	1.50	1847	0.25
	Urea + MSG	298	1.50	2040	0.89

20 Although the above NO_x reduction efficiency with urea was higher at the optimum temperature than the NO_x reduction efficiency with urea/monosodium glutamate, the temperature window for NO_x reduction was much wider with urea/monosodium glutamate. The addition of monosodium glutamate

25 significantly extended the reaction window to lower temperatures and enhanced the NO_x reduction efficiencies with urea. Close to 90% NO_x reduction was achieved at 1700°F with the addition of monosodium glutamate to urea.

30

Example 2

In this example, the N₂O reduction efficiency of urea/monosodium glutamate and urea were compared over a given temperature range. A 1 x 10⁶ BTU/hour furnace fired with natural gas at 3.5% excess oxygen was used to conduct

35 the experiment. The initial NO_x concentration (NO_i) in the combustion effluents ranged from about 297 to about 303 ppm. The molar ratio of nitrogen in reducing agent to NO_i was 1.5. The temperature decay versus time in the reaction zone was approximately 400°F/second. The experimental

results are r produced in Table 2 and shown graphically in Figure 3.

Table 2

	<u>Chemical</u>	<u>NO_i (ppm)</u>	<u>N/NO_i</u>	<u>Peak Temp. (°F)</u>	<u>N₂O (ppm)</u>
5	Urea	298	1.50	1454	1
	Urea	299	1.50	1614	4.5
10	Urea	303	1.50	1720	32
	Urea	297	1.50	1847	40
	Urea	298	1.50	2040	1
<hr/>					
15	Urea + MSG	298	1.50	1454	7
	Urea + MSG	299	1.50	1614	4
	Urea + MSG	303	1.50	1720	3
	Urea + MSG	297	1.50	1847	2
20	Urea + MSG	298	1.50	2040	0.5

The results of Example 2 confirm that the efficiency of N₂O reduction with urea depends strongly on the injection temperature, with the optimum near 1800°F. Use of urea/monosodium glutamate significantly eliminated the formation of N₂O emissions. Use of urea/monosodium glutamate also widens the temperature window for effective N₂O reduction. Using a 4:1 molar ratio of urea:monosodium glutamate, it is possible to achieve N₂O emissions in the range from about 0 ppm to about 8 ppm in a window between about 1450°F to about 2050°F.

Example 3

In this example, the NO_x reduction efficiency (NO_i/NO_f) of urea/sodium-containing compounds other than monosodium glutamate and urea were compared over a given temperature range. A 1 x 10⁶ BTU/hour furnace fired with natural gas at 3.5% excess oxygen was used to conduct the experiment. The

initial NO_x concentration (" NO_i ") in the combustion effluents ranged from about 298 to about 301 ppm. The ratio of reducing agent to NO_i was 1.5. The temperature decay versus time in the reaction zone was approximately 400°F/second. The experimental results are reproduced in Table 3.

Table 3

	<u>Chemical</u>	<u>NO_i, ppm</u>	<u>N/NO_i</u>	<u>Peak Temp., °F</u>	<u>NO_r/NO_i</u>
	Urea	298	1.50	1454	1.0
	Urea	299	1.50	1614	0.86
	Urea	303	1.50	1720	0.36
	Urea	297	1.50	1847	0.32
	Urea	298	1.50	2040	0.83
	Urea + NaOH	300	1.50	1850	0.27
	Urea + NaOH	300	1.50	1740	0.23
	Urea + Na_2CO_3	300	1.50	1850	0.27
	Urea + Na_2CO_3	301	1.50	1840	0.23

Similar to Example 1, the temperature window for NO_x reduction was much wider with urea/NaOH or Na_2CO_3 than with pure urea. The addition of these sodium-containing compounds significantly extended the reaction window to lower temperatures and enhanced the NO_x reduction efficiencies with urea.

Example 4

In this example, the N_2O reduction efficiency of urea/sodium-containing compounds (other than monosodium glutamate) and urea were compared over a given temperature range. A 1×10^6 BTU/hour furnace fired with natural gas at 3.5% excess oxygen was used to conduct the experiment. The

initial NO_x concentration (NO_i) in the combustion effluents ranged from about 297 to about 303 ppm. The molar ratio of nitrogen in reducing agent to NO_i was 1.5. The temperature decay versus time in the reaction zone was approximately 400°F/second. The experimental results are reproduced in Table 4.

Table 4

	<u>Chemical</u>	<u>NO_i (ppm)</u>	<u>N/NO_i</u>	<u>Peak Temp. (°F)</u>	<u>N_2O (ppm)</u>
	Urea	298	1.50	1454	1
	Urea	299	1.50	1614	4.50
	Urea	303	1.50	1720	32
	Urea	297	1.50	1847	40
	Urea	298	1.50	2040	1
	Urea + NaOH	300	1.5	1850	0
	Urea + Na_2CO_3	301	1.5	1850	0

The results of Example 4 confirm that the use of simple sodium-containing compounds, such as NaOH and Na_2CO_3 , significantly eliminate the formation of N_2O emissions. Using a 9:1 molar ratio of urea:sodium-containing compounds, it is possible to eliminate N_2O emissions with injections at 1850°F.

Example 5

An experiment was conducted similar to Example 2 with the exception that a comparison is made between the N_2O emission of urea/ $\text{Mg}(\text{CH}_3\text{COO})_2$ or $\text{Ca}(\text{CH}_3\text{COO})_2$ and urea over a given temperature range. The experimental results are reproduced in Table 5.

Table 5

	<u>Chemical</u>	<u>NO_i (ppm)</u>	<u>N/NO_i</u>	<u>Peak Temp. (°F)</u>	<u>N₂O (ppm)</u>
5	Urea	298	1.50	1454	1
	Urea	299	1.50	1614	4.50
	Urea	303	1.50	1720	32
	Urea	297	1.50	1847	40
	Urea	298	1.50	2040	1
10	<hr/>				
	Urea + Mg(CH ₃ COO) ₂	300	1.5	1850	16
	Urea+Ca(CH ₃ COO) ₂	301	1.5	1850	12

15 Similar to Example 2, the results of Example 5 confirm that the use of urea/Mg(CH₃COO)₂ or Ca(CH₃COO)₂ significantly eliminated the formation of N₂O emissions. The additions of 1200 ppm (by volume) of Mg(CH₃COO)₂ or Ca(CH₃COO)₂ to urea resulted in N₂O reductions of about 60% and 70% respectively.

20

Example 6

 An experiment was conducted similar to Example 2 except that a comparison is made between the N₂O emission of urea/Mg(CH₃COO)₂ and Na₂CO₃ and urea over a given temperature range. The experimental results are reproduced in Table 6.

25

Table 6

	<u>Chemical</u>	<u>NO_i (ppm)</u>	<u>N/NO_i</u>	<u>Peak Temp. (°F)</u>	<u>N₂O (ppm)</u>
30	Urea	298	1.50	1454	1
	Urea	299	1.50	1614	4.50
	Urea	303	1.50	1720	32
35	Urea	297	1.50	1847	40
	Urea	298	1.50	2040	1
	<hr/>				
40	Urea+Mg+Na ₂ CO ₃	300	1.5	1850	0

Similar to Example 2, the results of Example 6 confirm that the use of urea/Mg(CH₃COO)₂ and Na₂CO₃ significantly eliminated the formation of N₂O emissions. The additions of about 20 ppm Na₂CO₃ to the mixture of urea and Mg(CH₃COO)₂ completely eliminated N₂O emissions.

Example 7

An experiment was conducted similar to Example 2 to compare the N₂O emission of urea/NaOH or Na₂CO₃ and urea over a given temperature range. This example is similar to Example 4 with the exception that different concentrations of NaOH or Na₂CO₃ were employed. The experimental results are reproduced in Table 7.

Table 7

	<u>NO_x ppm</u>	<u>N/NO_x</u>	<u>Temp°F</u>	<u>N₂O ppm</u>
Urea	300	1.50	1740	40
Urea+NaOH(5-ppmNa)	300	1.50	1740	4
Urea+NaOH(20-ppmNa)	300	1.50	1740	1
Urea+Na ₂ CO ₃ (20-ppm Na)	300	1.50	1740	0

Example 8

Under the same furnace conditions as in Example 1, experiments were also conducted with the promoter injected separately from the urea. With the aqueous urea solution injected at 1850°F, an aqueous solution of Na₂CO₃, equivalent to 100 ppm sodium by volume, was injected at 2100, 1700, and 1550°F, separately from the urea. As shown in Table 8, all but the 1550°F injection case suppressed the formation of N₂O completely; 63% of N₂O was removed with the injection of Na₂CO₃ at 1550°F.

25

Table 8

		<u>NO_i ppm</u>	<u>N/NO_i</u>	<u>Na₂CO₃ Temp °F</u>	<u>N₂O ppm</u>
	Urea @ 1850°F	300	1.50	NA	49
5	Urea @ 1850°F	300	1.50	2100	0
	Urea @ 1850°F	300	1.50	1700	0
	Urea @ 1850°F	300	1.50	1550	18

10

Example 9

Similar to Example 8, experiments were also carried out to study the control of N₂O emissions produced by sources other than urea, including combustion systems such as fluidized bed combustors or from other chemical processes. Under the same furnace conditions, N₂O gas was introduced at 1800°F to simulate the formation of N₂O from fluidized bed combustors. Aqueous sodium carbonate solution was injected at 2150°F for the removal of N₂O emissions. It can be seen from Figure 4 that the efficiency of N₂O reduction with sodium carbonate depended upon the initial N₂O concentration and the Na to N₂O molar ratio. Fifty percent reduction could be achieved at a sodium to N₂O ratio of 0.1.

25

The data presented in Figure 4 is also documented in Table 9.

Table 9

SODIUM injected at 2150.°F
N₂O injected at 1800.°F

5	REDUCTION	N ₂ O	SODIUM/N ₂ O	N ₂ O
		(PPM)	Molar Ratio	(%)
10		93	0.21	61.4
		93	1.07	82.1
		93	1.61	94.0
15		600	0.03	33.8
		600	0.16	57.1
		600	0.33	74.0
		1350	0.03	35.0
		1350	0.14	61.0
		1350	0.16	65.0

20

Example 10

An experiment is conducted similar to Example 1 with the exception that the SO_x reduction efficiency of limestone (CaCO₃) is determined. It is found that the addition of alkali-metal compounds to limestone significantly enhanced the SO_x reduction efficiencies. Figure 5 presents these results.

From the foregoing, it will be appreciated that the present invention provides methods for controlling N₂O emissions from stationary combustion systems having variable flue gas temperatures.

It will also be appreciated that the present invention could provide methods for controlling N₂O emissions from stationary combustion systems while enlarging the useful temperature window and enhancing NO_x reduction.

It will be further appreciated that the present invention provides methods for controlling N₂O emissions from stationary combustion systems which allows one to treat flue gas with ammonia or ammonia precursors to reduce

NO_x emissions without producing an alternate class of pollutants such as N₂O.

5 It will be still further appreciated that the present invention provides methods for controlling N₂O emissions from stationary combustion systems while enhancing SO_x reduction. It will be still further appreciated that the present invention provides methods for controlling N₂O emissions from stationary combustion systems while enlarging the useful temperature window for NO_x and SO_x reduction.

10 It will be still further appreciated that the present invention provides methods for controlling N₂O emissions from stationary combustion systems which are economically feasible.

15 It will be still further appreciated that the present invention provides methods for controlling N₂O emissions from stationary combustion systems which perform effectively.

20 The invention may be embodied in other specific forms without departing from its spirit or essential characteristics. The described embodiments are to be considered in all respects only as illustrative and not restrictive. The scope of the invention is, therefore, indicated by the appended claims rather than by the foregoing description. All changes which come within the meaning and range of equivalency of the claims are to be embraced within their scope.

25 What is claimed is:

1. A process for controlling N_2O in effluent streams having temperature variations and containing N_2O and excess oxygen, comprising the step of introducing an N_2O control agent into the effluent stream at a point where the temperature of the effluent stream is in the range from about 1300 °F to about 3000 °F, said N_2O control agent being an alkaline compound of lithium, sodium, potassium, rubidium, cesium, francium, magnesium or calcium.

2. A process for controlling N_2O in effluent streams as defined in Claim 1, having temperature variations and containing N_2O and excess oxygen, comprising the step of introducing an N_2O control agent into the effluent stream at a point wherein the temperature of the effluent stream is in the range from about 1800°F to about 2800°F.

3. A process for controlling N_2O in effluent streams having temperature variations and containing N_2O and excess oxygen as in Claim 1, wherein the N_2O control agent is introduced to the effluent stream to control N_2O produced by the reaction of an NO_x reducing agent in the effluent stream, said NO_x reducing agent being ammonia and/or an ammonia precursor.

4. A process for controlling N_2O in effluent streams as defined in Claim 3, wherein the NO_x reducing agent is a compound selected from the group consisting of ammonia, urea, ammonium oxalate, ammonium acetate, ammonium formate, ammonium bicarbonate, ammonium carbonate, and ammonium sulfate.

5. A process for controlling N_2O in effluent streams as defined in Claim 1, wherein the N_2O control agent is an alkaline compound selected from the group consisting of alkaline-metal compounds of an organic acid, alkali-metal hydroxides, alkali-metal carbonates, and alkaline-earth metal compounds.

6. A process for controlling N_2O in effluent streams as defined in Claim 1, wherein the N_2O control agent is a compound selected from the group consisting of $NaOH$, Na_2SO_4 , $NaHSO_4$, Na_2CO_3 , monosodium glutamate, potassium glutamate, sodium formate, potassium formate, sodium acetate, potassium acetate, $LiOH$, $NaOH$, KOH , lithium carbonate, lithium bicarbonate, sodium carbonate, sodium bicarbonate, potassium carbonate, potassium bicarbonate, CaO , $CaCO_3$, $Ca(OH)_2$, $Ca(COOH)_2$, $Ca(CH_3COO)_2$, MgO , $MgCO_3$, $Mg(OH)_2$, $Mg(COOH)_2$, and $Mg(CH_3COO)_2$.

7. A process for controlling N_2O in effluent streams as defined in Claim 1, wherein the molar ratio of the alkali-metal or alkaline-earth metal in the control agent to N_2O in the effluent stream is in the range from about 0.05:1 to about 1.5:1.

8. A process for reducing NO_x while controlling the emission of N_2O in effluent streams having temperature variations and containing NO_x and excess oxygen, comprising the steps of:

(a) introducing an NO_x reducing agent into an effluent stream at a point where the temperature of

the effluent stream is in the range from about 1300 °F to about 2000 °F, said NO_x reducing agent being ammonia and/or an ammonia precursor; and

(b) introducing an N₂O control agent into the effluent stream at a point where the temperature of the effluent stream is in the range from about 1300 °F to about 3000 °F, said N₂O control agent being an alkaline compound of lithium, sodium, potassium, rubidium, cesium, francium, magnesium or calcium.

9. A process for reducing NO_x while controlling the production of N₂O in effluent streams as defined in Claim 8, wherein the N₂O control agent is an alkaline compound selected from the group consisting of alkaline-metal compounds of an organic acid, alkali-metal hydroxides, alkali-metal carbonates, and alkaline-earth metal compounds.

10. A process for reducing NO_x while controlling the production of N₂O in effluent streams as defined in Claim 8, wherein the N₂O control agent is a compound selected from the group consisting of NaOH, Na₂SO₄, NaHSO₄, Na₂CO₃, monosodium glutamate, potassium glutamate, sodium formate, potassium formate, sodium acetate, potassium acetate, LiOH, NaOH, KOH, lithium carbonate, lithium bicarbonate, sodium carbonate, sodium bicarbonate, potassium carbonate, potassium bicarbonate, CaO, CaCO₃, Ca(OH)₂, Ca(COOH)₂, Ca(CH₃COO)₂, MgO, MgCO₃, Mg(OH)₂, Mg(COOH)₂, and Mg(CH₃COO)₂.

11. A process for reducing NO_x while controlling the production of N_2O in effluent streams as defined in Claim 8, wherein the NO_x reducing agent is a compound selected from the group consisting of ammonia, urea, ammonium sulfate, ammonium carbonate, ammonium bicarbonate, ammonium formate, ammonium acetate, and ammonium oxalate.

12. A process for reducing NO_x while controlling the production of N_2O in effluent streams as defined in Claim 8, wherein the molar ratio of nitrogen in the NO_x reducing agent to the NO_x in the effluent stream is in the range from about 1:1 to about 3:1.

13. A process for reducing SO_x while controlling the production of N_2O in effluent streams containing SO_x and excess oxygen, comprising the steps of:

(a) introducing a SO_x reducing agent into an effluent stream at a point where the temperature of the effluent stream is in the range from about 800 °F to about 3000 °F, said SO_x reducing agent being an alkaline-earth metal compound; and

(b) introducing a N_2O control agent into the effluent stream at a point where the temperature of the effluent stream is in the range from about 1300 °F to about 3000 °F, said N_2O control agent being an alkaline compound of lithium, sodium, potassium, rubidium, cesium, francium, magnesium or calcium.

14. A process for reducing SO_x while controlling the production of N_2O in effluent streams as defined in

Claim 13, wherein the N_2O control agent is an alkaline compound selected from the group consisting of alkaline-metal compounds of an organic acid, alkali-metal hydroxides, alkali-metal carbonates, and alkaline-earth metal compounds.

15. A process for reducing SO_x while controlling the production of N_2O in effluent streams as defined in Claim 13, where the N_2O control agent is a compound selected from the group consisting of $NaOH$, Na_2SO_4 , $NaHSO_4$, Na_2CO_3 , monosodium glutamate, potassium glutamate, sodium formate, potassium formate, sodium acetate, potassium acetate, $LiOH$, $NaOH$, KOH , lithium carbonate, lithium bicarbonate, sodium carbonate, sodium bicarbonate, potassium carbonate, potassium bicarbonate, CaO , $CaCO_3$, $Ca(OH)_2$, $Ca(COOH)_2$, $Ca(CH_3COO)_2$, MgO , $MgCO_3$, $Mg(OH)_2$, $Mg(COOH)_2$, and $Mg(CH_3COO)_2$.

16. A process for reducing SO_x while controlling the production of N_2O in effluent streams as defined in Claim 13, wherein the molar ratio of the alkaline-earth metal in the SO_x reducing agent to the SO_x in the effluent stream is in the range from about 0.5:1 to about 5:1.

17. A process for reducing NO_x and SO_x while controlling the production of N_2O in effluent streams having temperature variations and containing NO_x , SO_x , and excess oxygen, comprising the steps of:

(a) introducing an NO_x reducing agent into an effluent stream at a point where the temperature of the effluent stream is in the range from about 1300 °F

to about 2000 °F, said NO_x reducing agent being ammonia and/or an ammonia precursor;

(b) introducing an SO_x reducing agent into an effluent stream at a point where the temperature of the effluent stream is in the range from about 800 °F to about 3000 °F, said SO_x reducing agent being an alkaline-earth metal compound; and

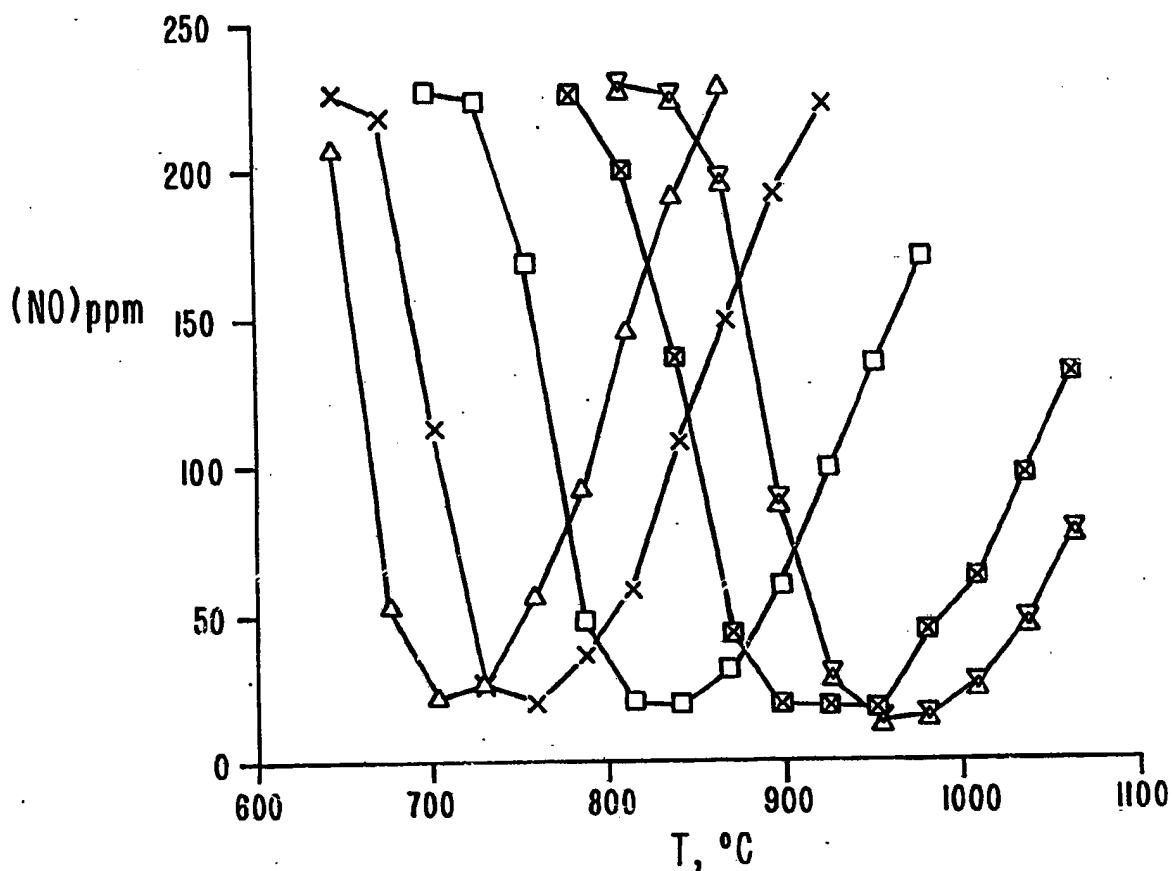
(c) introducing an N₂O control agent into the effluent stream at a point where the temperature of the effluent stream is in the range from about 1300 °F to about 3000 °F, said N₂O control agent being an alkaline compound selected from the group consisting of alkaline-metal compounds of an organic acid, alkali-metal hydroxides, alkali-metal carbonates, and alkaline-earth metal compounds.

18. A process for reducing NO_x and SO_x while controlling the production of N₂O in effluent streams as defined in Claim 17, wherein the NO_x reducing agent is a compound selected from the group consisting of ammonia, urea, ammonium sulfate, ammonium carbonate, ammonium bicarbonate, ammonium formate, ammonium acetate, and ammonium oxalate.

19. A process for reducing NO_x and SO_x while controlling the production of N₂O in effluent streams as defined in Claim 17, wherein the molar ratio of nitrogen in the NO_x reducing agent to the NO_x in the effluent stream is in the range from about 0.5:1 to about 5:1.

20. A process for reducing NO_x and SO_x while controlling the production of N_2O in effluent streams as defined in Claim 17, wherein the molar ratio of the alkaline-earth metal in the SO_x reducing agent to the SO_x in the effluent stream is in the range from about 0.5:1 to about 5:1.

1/5

Reduction of NO by H_2/NH_3 mixtures

$(NO)_0 = 225$ ppm, mass flow through reactor constant
and corresponding to 0.1s at 1038 °C

⊗ for 0 = H_2/NH_3

■ for 0.125

□ for 0.50

× for 1.3

△ for 2.4

$(NH_3)_0 / (NO)_0 = 2$

FIG. 1

2/5

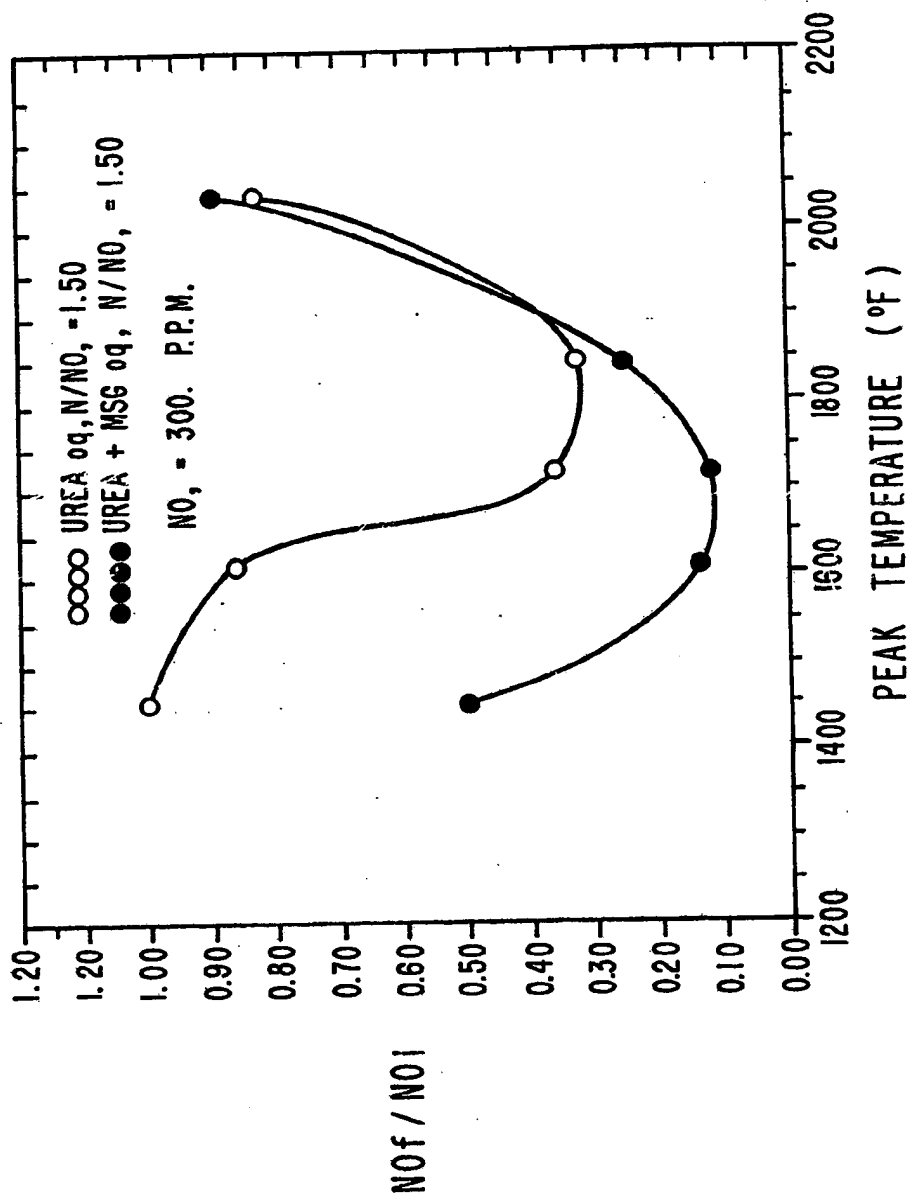


FIG. 2

3/5

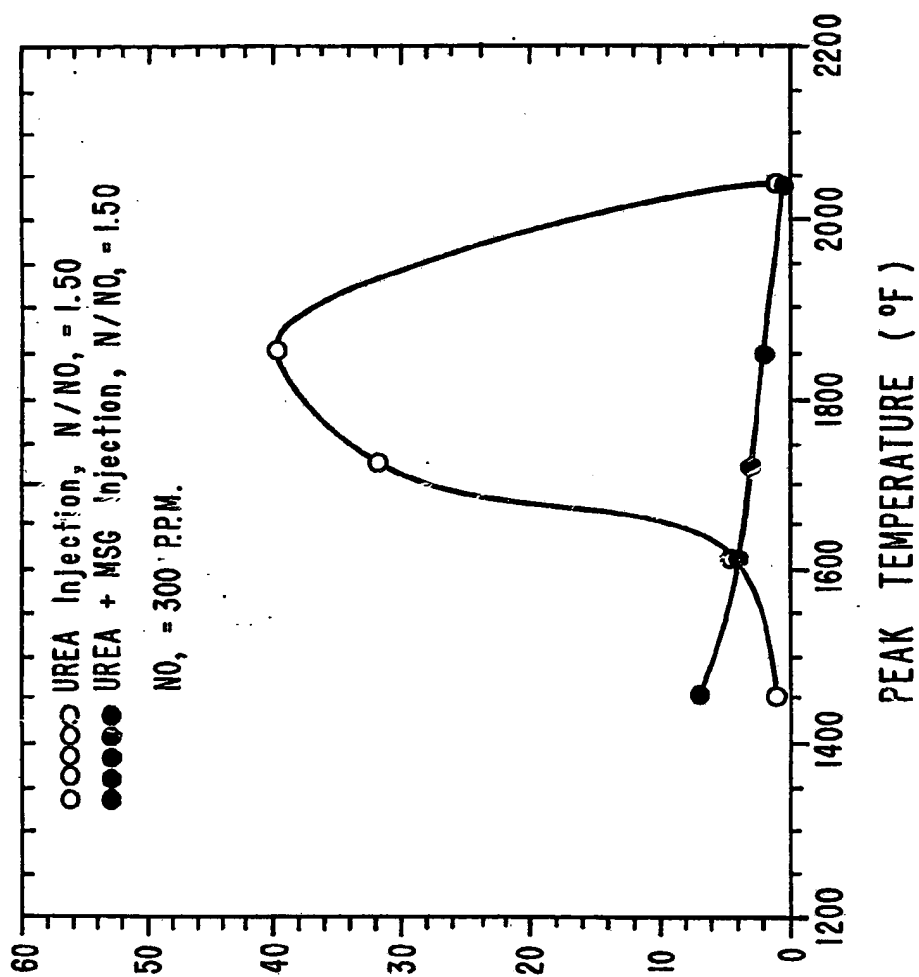


FIG. 3

4/5

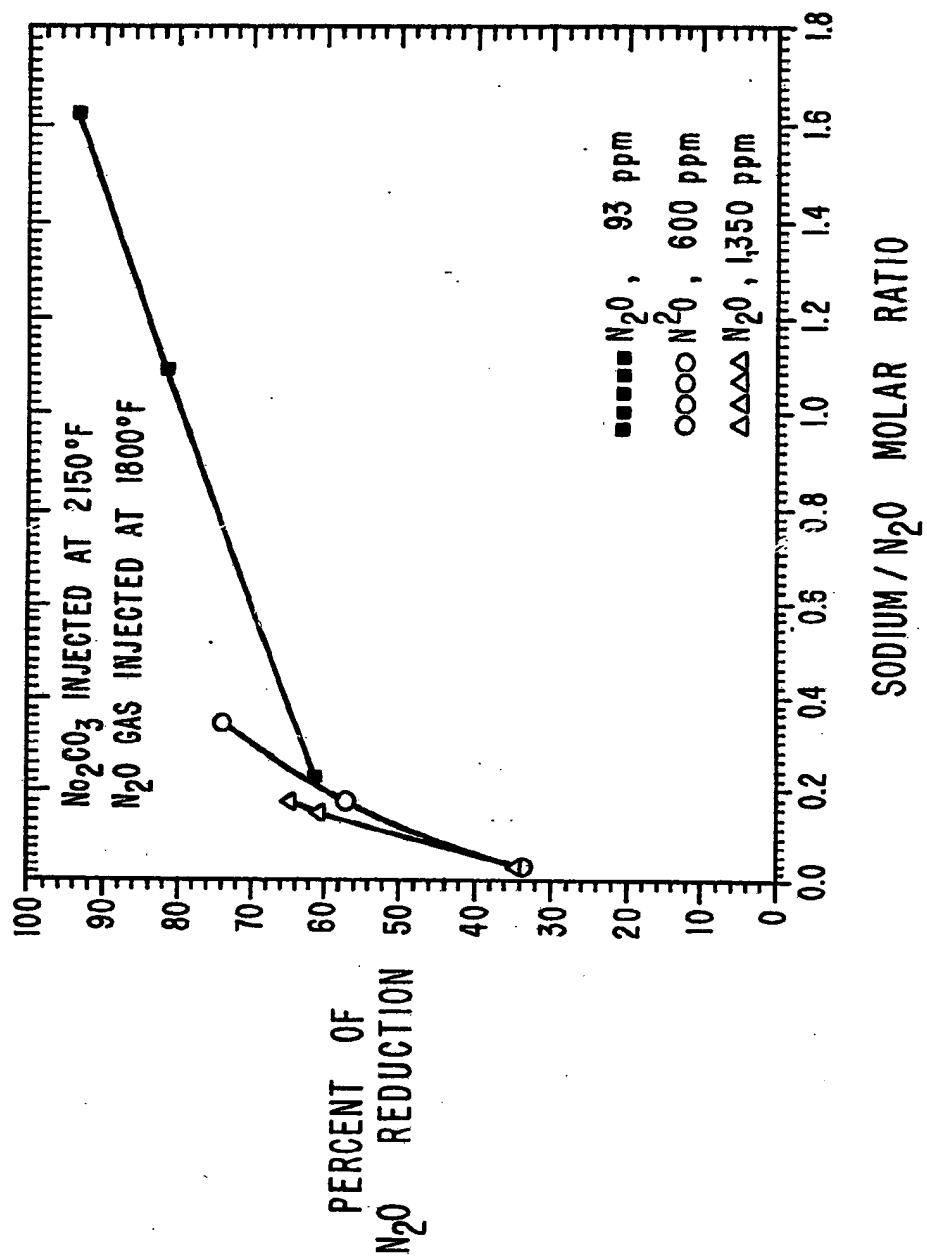


FIG. 4

5/5

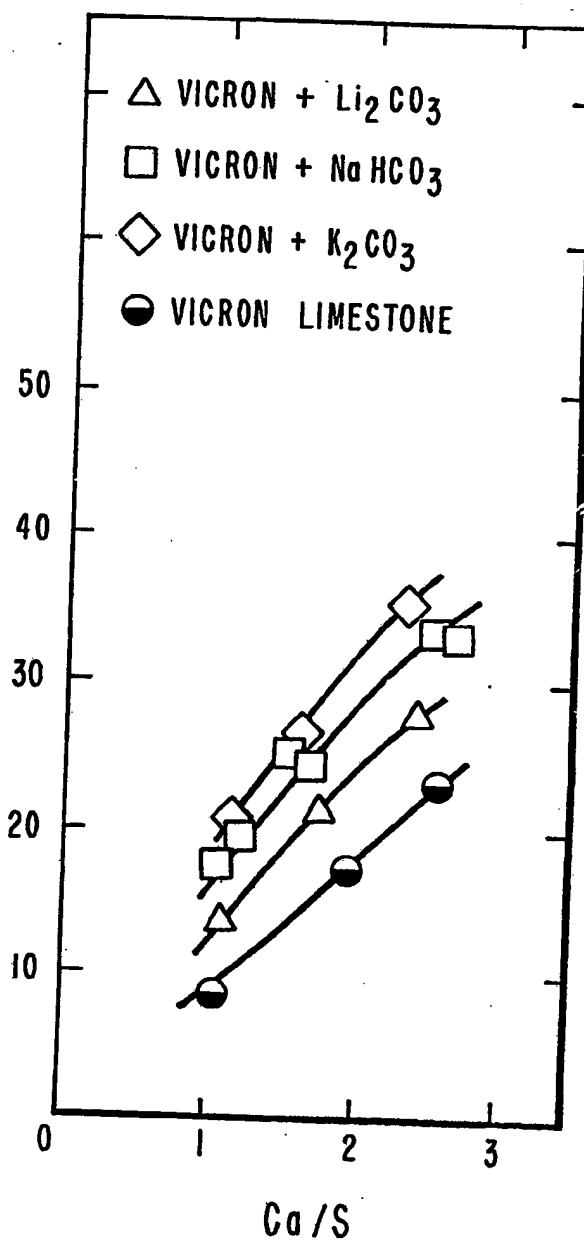
PERCENT
SULFUR CAPTURE

FIG. 5

INTERNATIONAL SEARCH REPORT

PCT/US93/02391

A. CLASSIFICATION OF SUBJECT MATTER

IPC(5) :C01B 21/00

US CL :423/235,351

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 423/235, 351

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US,A, 4,851,201 (HEAP ET AL) 25 JULY 1989	1-20
A	US,A, 4,861,567 (HEAP ET AL) 29 AUGUST 1989	1-20
A	US,A, 4,877,743 (WAUGH ET AL) 31 OCTOBER 1989	1-20
A	US,A, 5,048,432 (HOFMANN ET AL) 17 SEPTEMBER 1991	1-20

☐ Further documents are listed in the continuation of Box C.☐ See patent family annex.

* Special categories of cited documents:	T	later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
A document defining the general state of the art which is not considered to be part of particular relevance	*X*	document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
E earlier document published on or after the international filing date	*Y*	document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
L document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	*Z*	document member of the same patent family
O document referring to an oral disclosure, use, exhibition or other means		
P document published prior to the international filing date but later than the priority date claimed		

Date of the actual completion of the international search

18 MAY 1993

Date of mailing of the international search report

01 JUL 1993

Name and mailing address of the ISA/US
Commissioner of Patents and Trademarks
Box PCT
Washington, D.C. 20231

Facsimile No. NOT APPLICABLE

Authorized officer

GREGORY A. HELLER

Telephone No. (703) 308-2533